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A MOLECULAR DYNAMICS SIMULATION STUDY
OF LIQUID METAL TARGETS
USING THE EMBEDDED ATOM METHOD

by

Michael Lee Fisher

September 1988

Thesis Advisor:

Roger Smith

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A Molecular Dynamics Simulation Study
of Liquid Metal Targets
using the Embedded Atom Method

by

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ABSTRACT

A molecular dynamics digital simulation was used to investigate the sputtering yields from both liquid and solid metal targets. The system consisted of 1.0 keV Argon ions bombarding Rhodium targets. The embedded atom method of calculating potentials was used with a modified Moliere/Morse potential function. The yields from the solid and liquid targets were compared with the liquid showing a slightly higher yield than the solid. The liquid was simulated by random displacements of the atoms from a solid crystal lattice. Changing the seed, used by the random number generator to produce the liquid, effected the sputtering yield similar to moving the impact point. Four different sampling methods were investigated which produced similar results. Hence, the models described in this thesis should provide a basis for general sputtering simulations of liquids.



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I also wish to thank Professor Roger Smith, a colleague of Professor Harrison, who voluntarily assisted me in finishing this project.

I. INTRODUCTION

A. BACKGROUND ON SPUTTERING

Sputtering is the ejection of atoms or molecules from the surface of a target material as a result of the bombardment of that surface by particles. The general area of surface bombardment is usually divided into the two categories, of forward and backward effects. The formation of vacancies and interstitial atoms is an example of a forward effect which can occur; a vacancy and an interstitial atom together making up a Frenkel Pair. Scattering of the incident particles, ejection of electrons, and ejection of photons are just a few examples of the many backward processes that can occur.

The bombarding particles can be neutral or ionized atoms or molecules, neutrons, electrons, or photons. The ejected target particles can be neutral or ionized. In much of the research that has been done, the incident particles have been ions, and the target materials studied have been atoms, so that unless a specific differentiation is needed, the terms 'incident ions' and 'ejected atoms' will be commonly used.

In ion bombardment there are a number of important areas that require detailed study. In ion implantation the state of the impinging ion needs to be determined. How does it

lose its energy, what is its path, where does it come to rest? In dynamic recoil mixing, it is the state of the first atom hit by the impinging ion. This atom is called the primary knock-on atom (PKA). In the case where more than one atom is hit by the impinging ion, multiple PKA's can be produced. In sputtering, all the atoms of the target that are involved in the interaction, can potentially be ejected. These moving atoms form the collision cascade. The bombardment processes, both forward or backward, can be described by the motion of these atoms.

The quantitative measure of surface erosion by sputtering is the yield. The yield is defined as the number of ejected atoms per incident ion. Researchers have investigated the dependence of the yield on physical quantities such as the incident ion's energy, species, charge, and angle of incidence; and the target's composition and structure.

Sputtering can be categorized according to the microscopic mechanisms that cause the ejection of atoms from the target material. These include collisional, thermal, chemical, and electronic processes. Only collisional sputtering will be investigated in this project.

A bombardment begins as a collision between an incident ion and a target atom. If enough energy is transferred to the target atom, enabling it to overcome the binding energy of its lattice position, a recoil atom is created which can

collide with other target atoms causing a cascade. When the energy transferred back to a surface atom is greater than the binding energy at the surface, the atom is ejected. This energy is called the sputtering threshold.

The interaction that occurs between the ion and atom or between the recoil atom and subsequent target atoms can be described using the classical mechanics of two-body elastic collisions. If electronic energy loss is ignored, energy and momentum are conserved. We look specifically at the transfer of energy and momentum from the impinging ion to the atoms in the target. Inelastic energy losses such as electron excitation can occur as part of the dynamics, but consideration of these processes is beyond the scope of this project. In order to carry out the calculations it is necessary to specify the potential and the forces that occur in each collision [Refs. 1,2,3,4,5].

The interatomic potential function determines the action of the collision and the mechanics of the collision cascade. The potentials may be determined theoretically such as by performing Hartree Fock calculations or semi-empirically, that is, derived from experimental results. This method can be more useful because the theoretical models contain approximations which limit their accuracy. Among the potentials developed to date the screened Coulomb potential has been particularly successful. If there were no electrons around the nuclei of atoms the potential between

two atoms would be a simple Coulomb type. The presence of electrons changes the potential making it more complicated. The screened Coulomb potential accounts for the electrons by introducing an additional factor called the screening function. Many variations of the screening function have been tried. They generally produce good results in one energy range, but poor results in other ranges. More than one potential can be pieced together with splines to form a continuous potential over the energy range of interest. These piecewise potentials usually produce good results in sputtering calculations [Ref. 6].

B. LIQUIDS

Much sputtering research to date has involved the bombardment of solids by inert gas ions. Solid targets can take the form of a single crystal, a polycrystal, or an amorphous structure. In a single crystal there is long range order, with each atom located at a lattice point. In an amorphous structure the atoms still maintain short range order between neighbors, but the long range order is eliminated.

Liquids are similar to the amorphous solid, in that they lack long range order. The thermal energy associated with the atoms of the liquid allows them to vibrate further from their equilibrium positions, thus permitting the possibility of atoms exchanging positions and not returning to their original equilibrium position. This weakening of the

crystal structure prevents any definition of a fixed location, and therefore the material has no rigid structure [Ref. 7].

C. HISTORICAL OVERVIEW OF LIQUID SPUTTERING

Since most sputtering research has been done with solids, the area of liquid sputtering is in a relative infancy. One contribution from solid target research which seems applicable to liquids is that for low energy ion bombardment, an important factor effecting the yield is the surface condition of the target.

In 1970, Krutenat and Panzera [Ref. 8] looked at the yields from sputtering liquid and solid tin with Ar^+ ions of energies from threshold to 1200 ev, and at temperatures close to the melting point. One expected difference between the two surfaces was that the solid would build up the damage from the ion bombardment, whereas the liquid would "heal" rapidly from previous bombardments, and thus present a similar target to all incident ions. An experimental measurement of the yields for both polycrystalline solid and liquid tin showed that for ions of energy up to about 400 ev, the solid had a higher yield than the liquid. At about 400 ev the yields were the same, and the liquid yield was higher above 400 ev. Also, the yield for a remelted solid target with an incident ion energy of about 475 ev produced a yield that was between the liquid and solid yields.

Hurst and Cooper [Ref. 9] did an experiment in which they measured the yield from an indium target bombarded with Ar^+ ions of energy 107 eV, as they raised the temperature of the target, causing it to melt. This gave them data in both the solid and liquid phases for the same ion energy. They also ran the experiment in reverse by lowering the temperature, thus changing the target from liquid to solid. They found that the liquid produced a higher yield than the solid at this energy which is close to threshold. They also ran the experiment varying the ion energy from 17 eV to 190 eV bombarding both solid and liquid targets at temperatures close to the melting point, and found that the liquid yield exceeded the solid yield for all energies, but that the yield curves had the same shape over this range. They inferred that the difference in the yields was caused by the difference in binding energies of the two surfaces, but the sputtering effects due to the surface condition were approximately the same.

In 1982 Dumke, et al. [Ref. 10] did an experiment in which they sputtered the solid and liquid phases of gallium, indium, and a eutectic gallium-indium alloy, using argon ions. They found no significant change in the sputtering yields between the solid and liquid phases of the pure elements. The solid alloy showed a surface layer of indium which dominated the sputtering until the surface layer was sputtered away and the bulk material reached. For the

liquid alloy, again the surface was mostly indium, even though the alloy was 83.5% gallium and 16.5% indium. The indium sputtering yield was much higher than for gallium, indicating that most sputtered atoms must come from the surface monolayer.

D. COMPUTER SIMULATION

Computer simulation of physical systems is now being accepted as a useful adjunct to the more traditional theoretical and experimental approaches. Many simulations of sputtering have been completed over the thirty years since computers first became available. Few simulations with liquid targets have been done to date.

D. Y. Lo, et al. [Ref. 11] investigated collision cascades in liquid Indium using a multiple interaction computer simulation code, SPUT1. The liquid indium target was melted from a face centered cubic (FCC) structure and heated to approximately 900 K. The calculations were performed using pair potentials. A Moliere potential was used for interactions between the ion and target atom. For interactions between two atoms in the target the simulation used a potential comprised of a Moliere core joined to a Morse well, with a cubic spline [Ref. 11]. Analysis of the results by energy and angle resolved techniques showed relatively good agreement with experimental results and the theoretical predictions of Garrison [Ref. 11]. They concluded that the angular distribution of atoms ejected

from the first layer showed qualitative agreement with experiment, and the quantitative disagreement was probably attributable to the inadequacy of using pair potentials in the calculations.

II. OBJECTIVES

Most of the sputtering research done to date has used the method of pair potentials to perform the calculations. First a potential function is chosen for calculating the potential between two atoms in the sample. To calculate the force on a given atom, all the atoms in the sample are paired with the atom located at the point in question, and the potential is calculated for each pairing. The summation of the potential over all pairings yields the total potential of the atom in question. By repeating this process, the potential of every atom in the sample can be determined. From the potential the force can be derived.

The pair potential method has limitations on its applicability to certain physical systems. This method gives good results for the internal collision cascades, but the calculation is less accurate in dealing with surface phenomena [Ref. 12].

A new method of calculating the potential of a given atom in a sample was proposed in 1983. This new method, called the embedded atom method (EAM), was first proposed by Daw and Baskes [Ref. 12]. In the embedded atom method all atoms in the sample are viewed as being embedded in the host consisting of all other atoms. The energy depends on the electron density. This overcomes some of the problems

associated with large volume dependent terms inherent in the pair potential approach. The semi-empirical nature of the embedded atom approach allows the potentials to be determined in terms of well defined and measurable quantities.

Both methods described above have been applied to liquid targets to a limited degree, but there is some doubt as to the validity of using the pair potential method to calculate the energetics of an amorphous target. On the other hand the EAM avoids some of the drawbacks of the pair potential method, and can still handle the lack of structure associated with a liquid [Ref. 13]. For this reason the EAM will be used for all the calculations in this project.

The main objective of this project is to make a direct comparison of the results produced by calculating the sputtering yield of an ion bombardment system in two different states, solid and liquid. The solid target will consist of a perfect crystal, while the liquid target will be the same crystal which has been warmed above its melting point. This analysis will allow conclusions to be drawn as to the effect of structure or long range order on a sputtering yield.

Different types of trajectory sets for the liquid targets will be used in order to investigate the effect of sampling methods on a liquid target system.

All of the calculations and results derived in this project will be accomplished using digital simulation techniques. In order to ensure a proper comparison, all calculations will be completed using the same target, with the exception that atoms in the liquid target will have been displaced from their crystal lattice positions, an amount which on average is equal to the mean thermal displacement. Such things as target element, size, impact points, etc. will remain fixed.

III. MODEL AND SIMULATION

A. MULTIPLE INTERACTION MOLECULAR DYNAMICS SIMULATION

The computer code used to model the ion bombardment simulation in this project was developed by Professor Don E. Harrison, Jr. at the Naval Postgraduate School. The program used in this project is called EWARM. It is an offshoot of predecessors called EDYN and QDYN. QDYN is the grandfather of all of them. It had been developed to do sputtering simulation research using pair potentials. EDYN is similar to QDYN except that it uses the EAM to calculate the forces needed for the simulation. A detailed account of the digital simulation method, the mathematical model and the operation of the integration technique has been previously published by Harrison, et al. [Refs. 14, 15, 16, 17, 18, 19].

EWARM differs from the other two in that it contains the ability to handle the simulation of a liquid target. The major difference is in the addition of a WARMER subroutine which randomly dislocates the atoms of the target from their perfect crystal lattice sites, thus producing an amorphous target.

All three of these programs handle the molecular dynamics in the same way. EDYN is completely contained within EWARM. EWARM is a continuous time simulation of

sputtering, which means that the program proceeds through a series of timesteps. Each timestep consists of a calculation of the forces, the new velocities and the positions of each atom, at the end of the timestep, movement of the atoms to their new positions, and a test for energy conservation. Both programs compute the atoms' trajectories by performing a numerical solution of Newton's classical equations of motion. The length of subsequent timesteps is controlled by a predictor-corrector integration scheme which determines the time increment from the fastest moving particle [Ref. 17].

The system used for this project consists of an impinging argon ion and a target of 912 rhodium atoms. Each particle in the system is characterized by mass, position, velocity, and the force laws by which it interacts with other particles.

After the impinging ion hits the target at the specified impact point, the trajectories of all the particles in the system develop in time as the energy and momentum are dissipated through the target. The calculation is called a trajectory and the dissipation through the target is called a collision cascade. The trajectory is terminated when the energy and momentum has dissipated to the point where no more atoms will be ejected. In this particular system the calculation ends when the maximum kinetic energy of a single particle falls below 2.5 eV.

B. THE EMBEDDED-ATOM METHOD

In the embedded-atom method each atom is treated as if it were embedded in the host of all other atoms. The energy required to embed an atom depends on the electron density. This electron density can always be defined, so any problems associated with not being able to define a volume are avoided. Therefore the embedded atom method can be used on targets with surfaces, and promises better results when applied to liquid targets. Another important factor is that the embedded-atom method is not significantly more complicated to use than pair potentials [Ref. 12].

To begin we think of each atom as an impurity embedded in the host material. The total potential energy is the sum of the host and impurity contributions. The host potential is a function of the electron density without the impurity, and the impurity potential depends on the position and charge of the impurity nucleus. Therefore the energy of the host with impurity is a functional of the host electron density and a function of the impurity type and position. The functional is universal, independent of the host material. Its form is not known, but a simple approximation assumes that energy only depends on the other atoms that are close to the impurity, or the impurity experiences a locally uniform electron density. With this approximation the functional of the electron density becomes a function and only the electron density at the impurity position is

needed. This makes the position dependence trivial and the energy depends only on the electron density of the host at a given point plus an electrostatic interaction. The total energy is a sum over all contributions from the individual atoms. The electrostatic interaction and the electron density terms are determined empirically [Refs. 10, 12]. Daw and Baskes have previously published articles describing the approximations and details of the derivation of the embedded atom method [Refs. 10, 12].

A new version of QDYN was created using the embedded-atom method to calculate the potentials of the atoms in the system. Called EDYN, it employs a method whereby the total potential energy of a given atom is the sum of the short-range pairwise potential energy and the embedding energy.

C. WARMING THE TARGET

The next modification implemented in QDYN and EDYN was the addition of the subroutine WARMER. This routine takes a perfect crystal lattice and randomly moves the atoms so that the resulting target is amorphous. Specifically, this is done by adding a correction term to each component of the initial positions of the perfect crystal target atoms. This correction term is simply a normally distributed random variant multiplied by a thermal amplitude factor. The temperature can be approximated from the change in the potential energy of the system so that the target can be "warmed" to a desired temperature. If the target is warmed

to the melting temperature of the material, then the resulting target is a liquid.

Since the warmer randomly locates the atoms of the target, it needs a supply of random numbers. These come from a subroutine that generates pseudo-random numbers. As with most pseudo-random number generators, the user has control of the starting point of the generator through the seed. Each number produced by the generator is then used as the seed for the next number. Throughout this project the starting and ending seeds are monitored so that a calculation can be repeated by using the same starting seed.

These starting seeds are very important because changing the seed will change the way in which the target atoms are dislocated, and therefore produce a different target. For example, to run a set of trajectories with the same target would require that the same starting seed be used for the warming of each trajectory target. This line of thinking can lead to many possible ways of warming a target for a set of trajectories.

D. POTENTIAL FUNCTIONS

In a simulation of this type, the interatomic potential functions must be chosen so that the forces between atoms can be calculated. These functions ultimately control the dynamics of the simulation. Approximations will occur, because of a lack of detailed knowledge in certain regions of the interatomic potentials. At separations much greater

than the equilibrium separation of two atoms, there is relatively good understanding of the potentials. Likewise, at the equilibrium separation, but at the separation distances less than equilibrium, and in the transition area where the separation distance is just greater than equilibrium, much less is known.

There are several theories available as well as some empirical methods for obtaining potential functions to use in a calculation. Most of these potential functions take the form of a "wall" or almost vertical slope as the interatomic distance approaches zero, decreasing to a "well" or minimum at the equilibrium separation, increasing through a steady transition approaching a horizontal line far from the equilibrium separation distance. This form of a potential function will produce an attractive force at distances greater than the equilibrium separation, zero force at equilibrium, and a repulsive force which gets very large in the area where the separation distance approaches zero [Ref. 6].

In most cases, one function does not describe the interatomic potential very well over all of the regions described above. Several functions are generally splined together to produce a composite function which is good over all regions, and can be verified by existing experimental data.

In this project the interatomic potentials are empirical. They were created from several functional forms which agree with experimental data in the region where they are employed. They are functions that use certain constants related to physical parameters of the atoms involved.

The functions used in this project to describe the interatomic potentials can be divided into two types. The first describes the interactions between the ion and a target atom (Ar-Rh), and the second describes the interactions between two target atoms (Rh-Rh).

The Rh-Rh function consists of a modified Moliere potential joined to an attractive Morse potential using a cubic spline. This is given by:

$$V = [(Z_1 Z_2 e^2 / ka) / (R / ka)] g(R / ka) \quad R < R_a$$

$$g(R / ka) = [0.35 \exp(-0.3 R / ka) + 0.55 \exp(-1.2 R / ka) + 0.10 \exp(-6.0 R / ka)]$$

$$a = 0.8853 a_0 / [Z_1^{1/2} + Z_2^{1/2}]^{2/3}$$

$$V = C_0 + C_1 R + C_2 R^2 + C_3 R^3 \quad R_a < R < R_b$$

$$V = D(\exp[-2b(R - R_e)] - 2 \exp[-b(R - R_e)]) \quad R_b < R < R_c$$

$$V = 0 \quad R > R_c$$

where:

V is the potential

Z_1 is the atomic number of the first atom

Z_2 is the atomic number of the second atom

e is the charge of an electron

R is the distance between the two atoms

k is a magnitude factor between 0 and 1

a_0 is the Bohr radius (0.529 Å)
 g is the screening function
 C_0, C_1, C_2, C_3 are fitting parameters
 D is the well depth
 R_e is the equilibrium separation distance
 b is a fitting parameter (\AA^{-1})

In reality the potential function exists out to infinite separation. The composite function is truncated at R_c because beyond this point its effect is negligible [Ref. 6].

The potential function that was used for interactions between Ar^+ and Rh is an unmodified Moliere function of the form:

$$V = [(Z_1 Z_2 e^2 / a) / (R/a)] g(R/a) \quad R < R_a$$

$$V = 0 \quad R > R_a$$

The parameters that define the potential functions for all the runs generated in this simulation are given in table 1.

TABLE 1
POTENTIAL FUNCTION PARAMETERS

Parameter	Rh-Rh	Ar-Rh
D (ev)	0.7595	0.0
R_e (Å)	2.750	0.0
b (\AA^{-1})	1.080	0.0
R_a (LU)	0.73	1.70
R_b (LU)	0.80	1.70
R_c (LU)	2.05	1.70
k	0.74	1.0

E. PHYSICAL PARAMETERS

The target is produced by building a perfect crystal, and then "warming" the crystal to the melting point of rhodium (2239 K), thus producing an amorphous liquid. The target is oriented so that the (111) face of the perfect crystalline target would be normal to the incoming ion. This notation of a (111) face is not relevant for a liquid but does give a basis for comparison with results from the solid crystal.

Rhodium is a face centered cubic metal (fcc) with a lattice constant (a_0) of 3.804 Å. The basic unit of distance used is the lattice unit which is defined as half the interatomic spacing. For this simulation the lattice unit is equal to 1.902 Å.

In the simulations it is important to choose the target size judiciously. A target size must be chosen that will produce realistic results yet keep within the constraints of the computer resources available.

After extensive experience with these types of simulations, Harrison has developed a working definition of containment [Ref. 17]: "If an increase in target size does not change the results of the computation, the trajectory is effectively contained for the purposes of that computation."

As the target size increases, the observable will approach its absolute value asymptotically as full

containment is reached. In further clarification of this point Harrison states that:

As one might anticipate from experimental experience with absolute values, yield provides the most severe test of containment; so absolute yield containment guarantees everything else. As the target size increases, all other global results reach constant values for much smaller targets than those required to produce stable yield values [Ref. 17].

F. ENSEMBLES OF TRAJECTORIES

Another item requiring careful choice is the method by which the trajectories are chosen so that the result is an average of the observable that we wish to measure. A method has been developed for perfect crystal targets, and has also been applied to warmed targets [Ref. 19]. The method uses the concept of a representative area. Each impact point on the target surface produces a different trajectory. By symmetry, a subset of impact points on the target surface will produce every possible trajectory for a given lattice orientation. The representative area is this subset of impact points that contains every possible trajectory for a given target surface. Since there are an infinite number of possible trajectories, the sample size requires judicious choice. A carefully planned sample of trajectories over this representative area should produce the average value for the observable. The set of all possible trajectories is called an ensemble, and the smaller sample set is known as a sample ensemble. Regular sampling of the representative area using upwards of 300 impact points has been used very

successfully in the past and will be used in this simulation. A possible representative area is a rectangle for the (111) surface. A starting point, called a pinpoint is placed in the lower left corner of the representative area, and a set of impact points are regularly spaced from the pinpoint over the representative area [Ref. 19].

For this project a set of 300 impact points has been chosen. This set of impact points will be used for the perfect crystal target as well as for the liquid targets. The use of this sampling method has been substantiated previously for perfect crystal targets [Ref. 19], but when the target melts the justification for its use is more questionable. This project aims to determine a legitimate method of sampling for liquid targets.

G. ANALYSIS

The data from the simulation program can be analyzed using a fortran program called ANPLOT which takes the information about the individual ejected atoms and produces graphical presentations allowing comparison to other simulation or experimental results. It produces various plots and histograms such as atoms per single ion, ejected atom energy distribution, atom ejection time distribution, etc. This program was used to analyze all the data derived from this project.

IV. RESULTS

A. OVERVIEW

The results presented in this project were produced by a series of simulations. The total amount of computer time used to collect this information is in excess of 90 hours. A single sampling method was employed for the solid case, but six different methods were tried with liquid targets in order to investigate sampling methods to be used. Of the six methods tried, four appear to be useful for sputtering. In the solid case 300 trajectories were run with a perfect crystal target. The target was the same for each trajectory. The first liquid set consisted of 300 trajectories hitting a target that had been warmed. The target was identical for each trajectory. The next data set also consisted of 300 trajectories, but a different seed was used to produce the target for each trajectory, so that each trajectory was hitting a different target. Two sets of 100 trajectories were run in which the impact points remained fixed while the seed was changed, thus producing different targets for each trajectory.

For the last three sets, four atoms in the initial impact area were frozen in their crystal lattice positions while all the other atoms were randomly dislocated by the warmer. These four atoms enclose the representative area in

the perfect crystal target, and so it was postulated that by freezing them, some credence could be given to using this sampling technique on a liquid target. One set of 300 trajectories was run with the same warming seed for each trajectory, one set of 300 trajectories run with a different seed for each trajectory, and finally the last set of 100 trajectories was run keeping the impact point fixed and changing the seed for each trajectory.

The trajectory sets completed with fixed impact points were analyzed as distributions to determine the possibility of using this method as a sampling technique, and to see what kind of variation would be produced at a single impact point. From all the data it is apparent that changing the seed does effect the trajectory, and therefore the sputtering yield.

B. SPUTTERING YIELD

The total sputtering yields calculated for the four 300 trajectory sets using liquid targets were approximately the same. There is less than a 2% difference between any single set average and the average of the four. The fact that these four sets independently reproduced the same relative yield would indicate that the sample is probably large enough for this observable. It does not give any indication that any of the sampling methods is better than another, and this paper has not tried to optimize the sample size or method. The average of these four sets indicates that the

liquid target produced about a 6% higher yield than the solid target. The single impact point trajectory sets showed reproducibility, but did not agree with the other liquid sets. This means that for our method of warming the crystal, changing the impact point on the target is not the same as changing the target behind the impact point, probably due to the relatively small thermal displacements of the atoms from their initial positions. This should be explored further in any future work. The total sputtering yields are presented in table 2.

TABLE 2
TOTAL SPUTTERING YIELDS

Category	Atoms Ejected	Trajectories	Average Atoms per Trajectory
perfect crystal	1596	300	5.32
liquids:			
single target	1670	300	5.57
different targets	1731	300	5.77
liquids with 4 representative area atoms initially fixed			
single target	1706	300	5.69
different targets	1725	300	5.75
liquids with fixed impact point			
point 1	795	102	7.79
point 2	748	103	7.26
point 1*	803	108	7.44

* same impact point, except that four atoms in the representative area are fixed

Because of the apparent difference between the single impact point sets and the other liquid sets, the multiple impact point sets will be compared to the solid set in the remainder of the results. The single impact point sets are too locally controlled by the choice of impact point. The multiple impact point sets give a better sampling diversity.

C. ATOMS EJECTED PER SINGLE ION

Atoms per single ion (ASI) is an important quantity which, in sputtering simulations of solids can be determined relatively accurately using the molecular dynamics simulation method. The sputtering yield is the mean ASI over the sample set. The ASI distribution is difficult to obtain in an experiment, and simulations can often gain useful physical information. In this simulation the liquid ASI distributions did not differ significantly from the solid ASI distribution, except in producing a higher mean (Table 2). Figures 1 through 5 show the ASI distributions for the solid trajectory set and the four liquid trajectory sets. The two liquid distributions (Figures 2,3) that used the same target for all trajectories closely follow the solid distribution (Figure 1). The other two liquid targets that used a different seed for each trajectory (Figures 4,5) only differ slightly.

D. EJECTION TIMES

Figures 6 through 10 provide a view of the ejection time distributions for the five cases of interest. There is no

apparent difference in the ejection time distributions between the liquid and solid cases. Also, the sampling method appears to make little difference in the liquid cases. An important point is that about 95% of all the ejections of this system occur within first 200 femtoseconds of the simulation, much shorter than the time scale for thermal effects.

E. EJECTED ATOM ENERGY DISTRIBUTIONS

There is very little difference between any of the energy distributions of the ejected atoms, meaning that the state of the target does not effect the energy distribution of the sputtered particles. Also, the sampling method does not appear to effect the resulting energy distributions.

F. EJECTED ATOM ANGULAR DISTRIBUTIONS

Sputtering simulations can also determine the angular distributions of the atoms as they are ejected. This is a simulation observable that compares well with experimental sputtering data [Ref. 20]. A set of plots has been developed by Harrison to show the polar angular distributions. These distributions are created by tallying the ejected atoms into bins five degrees wide from 0 to 90 degrees. 0 degrees is normal to the surface. If the low energy atoms, say below five ev are excluded, the comparison to the experimental data is even better. In this project all of the ejected atoms were included in the distributions.

The distributions, presented as figures 11 through 15, show little difference between the liquid and solid cases. Also, there is no detectable difference among the four cases of the liquid. The peaks of these distributions all occur about 30 degrees.

By excluding those ejected atoms with less than 20 ev of energy, the distributions are skewed to smaller polar angles, meaning that the higher energy atoms must be ejected at angles closer to normal than those atoms with lower energy. Figure 16 is provided as a comparison for the solid case. The liquid distributions show the same trend.

G. SPOT PATTERNS

Spot patterns are another method of showing the angular distributions of the ejected atoms. Spot patterns have an advantage over the polar angular distribution plots, namely that the spot patterns present the azimuthal angular dependence simultaneously. Figures 17 through 19 show the spot patterns for the solid case and the four liquid cases. The solid case shows its distinctive six fold symmetry pattern. In the liquid cases this pattern is still evident, but not as sharp.

H. ATOM YIELD PER IMPACT POINT

Figures 20 through 24 are the graphical representations of the relative yields produced at each impact point for the five cases being studied. This displays each impact point on the target surface, and then indicates the relative yield

at that point by a series of tree rings. Higher yields are shown by more rings. The locations of the centers of the first surface layer target atoms have been plotted to give a better picture of where the atoms are relative to the impact points. In the three targets in which the seed was identical for each trajectory (Figures 20,21,22), the ejections were less uniformly distributed than for those in which the target was changed from trajectory to trajectory (Figures 23,24). In neither case were the ejections uniformly distributed over the representative area, which should be the case for a purely random target.

V. CONCLUSIONS

This project has made no attempt to compare the results of the simulations with experimental results. The comparisons reported, have been between similar systems in two different states, solid and liquid, and between identical models of liquid systems in which the sampling methods were varied.

All of the sampling techniques used produced about the same results for the calculated yields. Changing the seed to the random number generator changes the trajectory, as shown by the distributions produced at the same spatial impact point. Figure 25 is an example of the distributions that resulted from this sampling technique. The distributions for the other observables were similar. Changing the impact point produced better statistics than keeping the impact point fixed and changing the seed. This is because the thermal displacements were with respect to positions based on a crystal lattice. Keeping the impact point fixed maintains some of the underlying crystal structure, and hence the target is not truly randomized. Computer simulation of sputtering from liquids should not use this sampling technique.

Any of the other four methods should give reasonable statistics. It appears that the size of the representative

area chosen for solid simulations because of lattice symmetry also works well for the randomized target.

Another alternative is to use a stochastic sampling technique of the representative area, in addition to randomizing the target for each trajectory.

A possible improvement to the model would be to simulate a warmed target by adding thermal velocities in addition to displacements. Any initial kinetic energy an atom may have could affect its ejection probability. It is recommended that this approach be implemented in any future studies of liquid targets.

The effect of warming a target, even past its melting point, does not substantially change the calculated sputtering yields. A slight increase in yield occurs as a result of the warming, but the ejected particle distributions do not change.

The work described in this project used the embedded atom method to perform all the calculations. This is the new and promising method, but most current research still uses pair potential calculations. Other many body potentials have now been reported for modelling equilibrium phenomena [Ref. 21] which promise to give even better models of the underlying physics. An extension of this project would be to run the simulations using a variety of potentials, and to compare with results obtained by experiment.

Further study of different sampling techniques would only improve the confidence in any sampling method ultimately selected.

APPENDIX

1.0 RH(111)/AR<111> SOLID TARGET
ATOMS/PER SINGLE ION

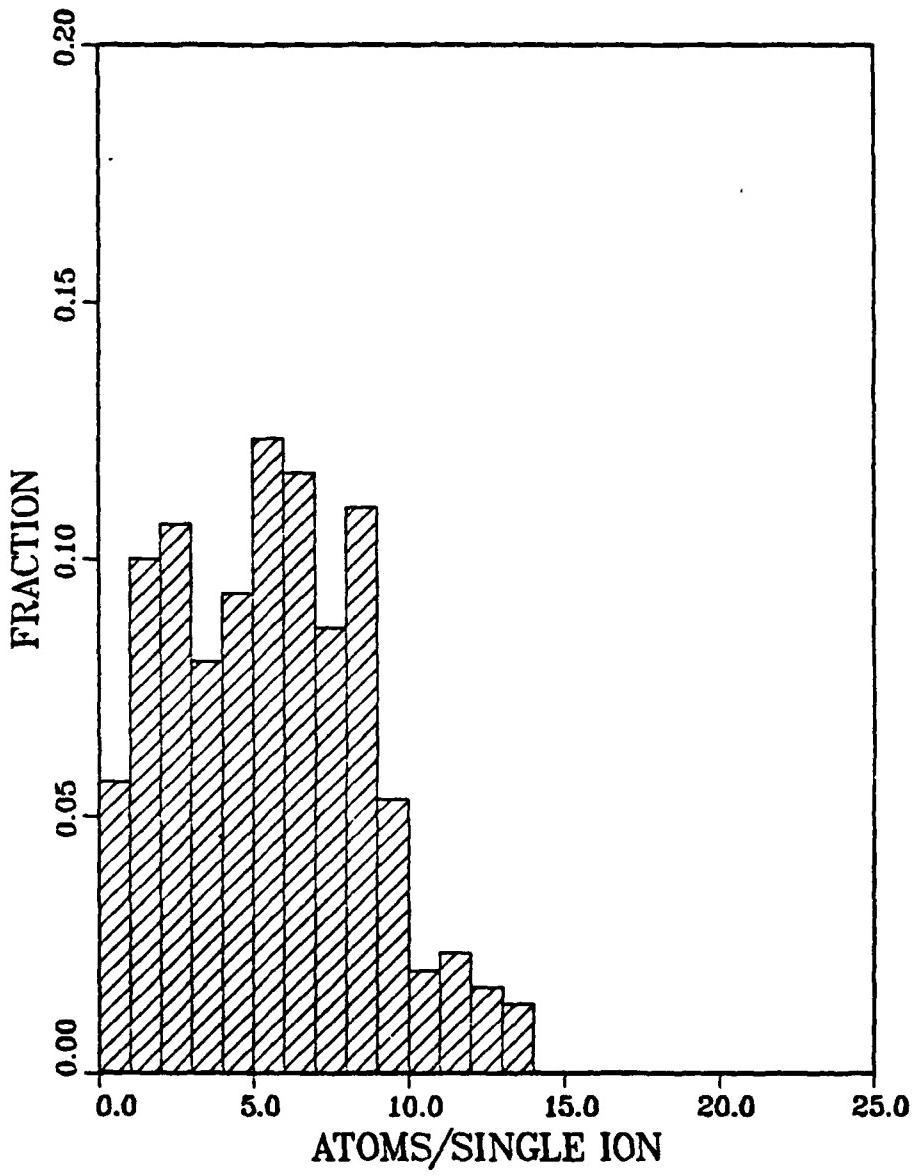


Figure 1 Atoms per Single Ion Distribution.

1.0 RH(111)/AR<111> WARMED SINGLE TARGET
ATOMS/PER SINGLE ION

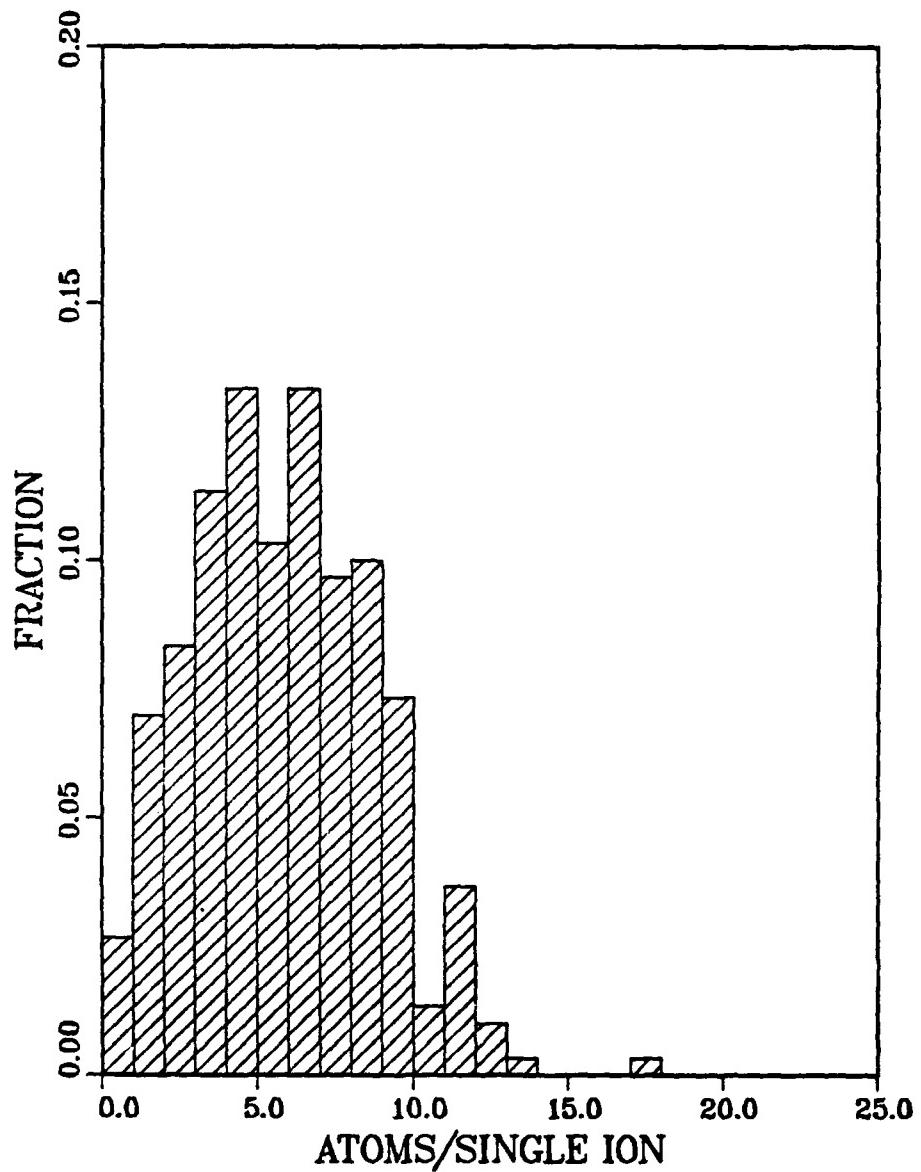


Figure 2 Atoms per Single Ion Distribution.

1.0 RH(111)/AR<111> WRMD SING TARG W/4 FROZEN
ATOMS/PER SINGLE ION

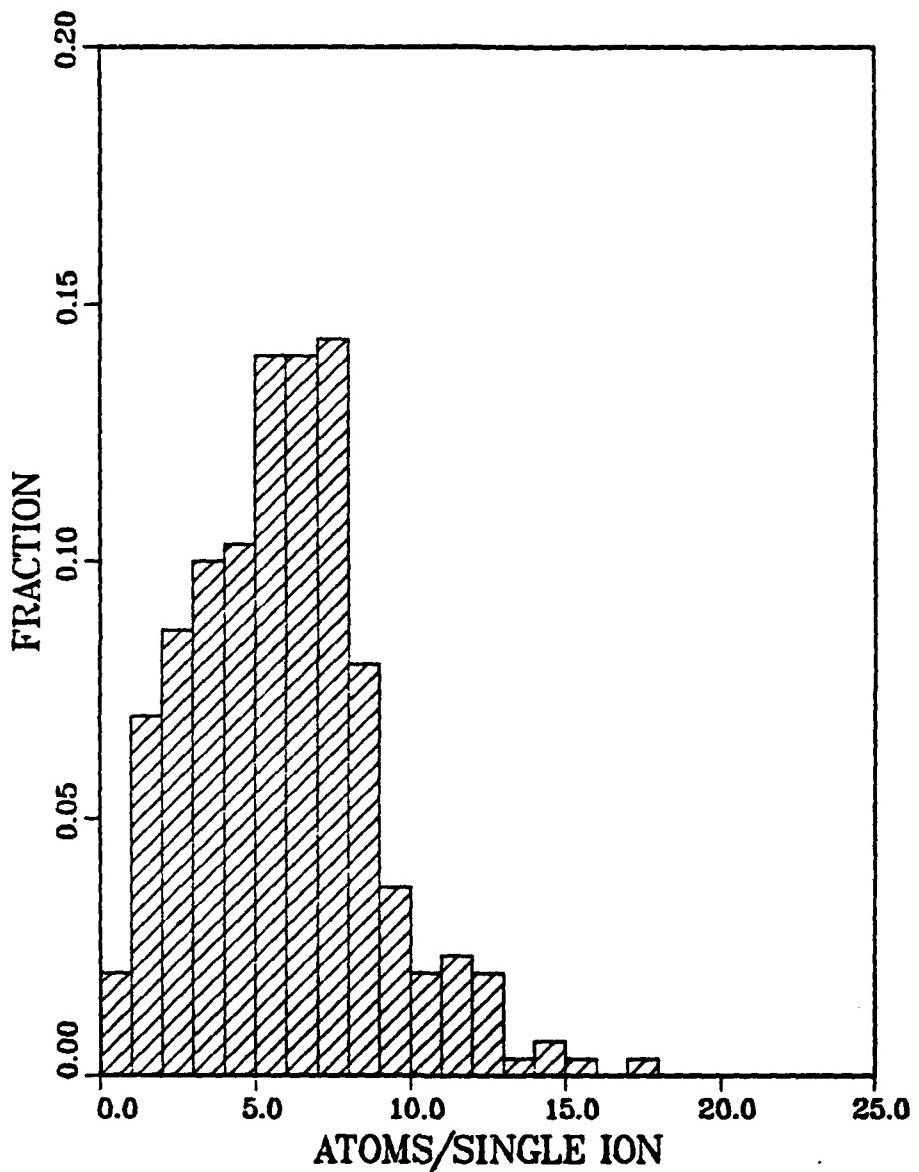


Figure 3 Atoms per Single Ion Distribution.

1.0 RH(111)/AR<111> WARMED DIFFERENT TARGET
ATOMS/PER SINGLE ION

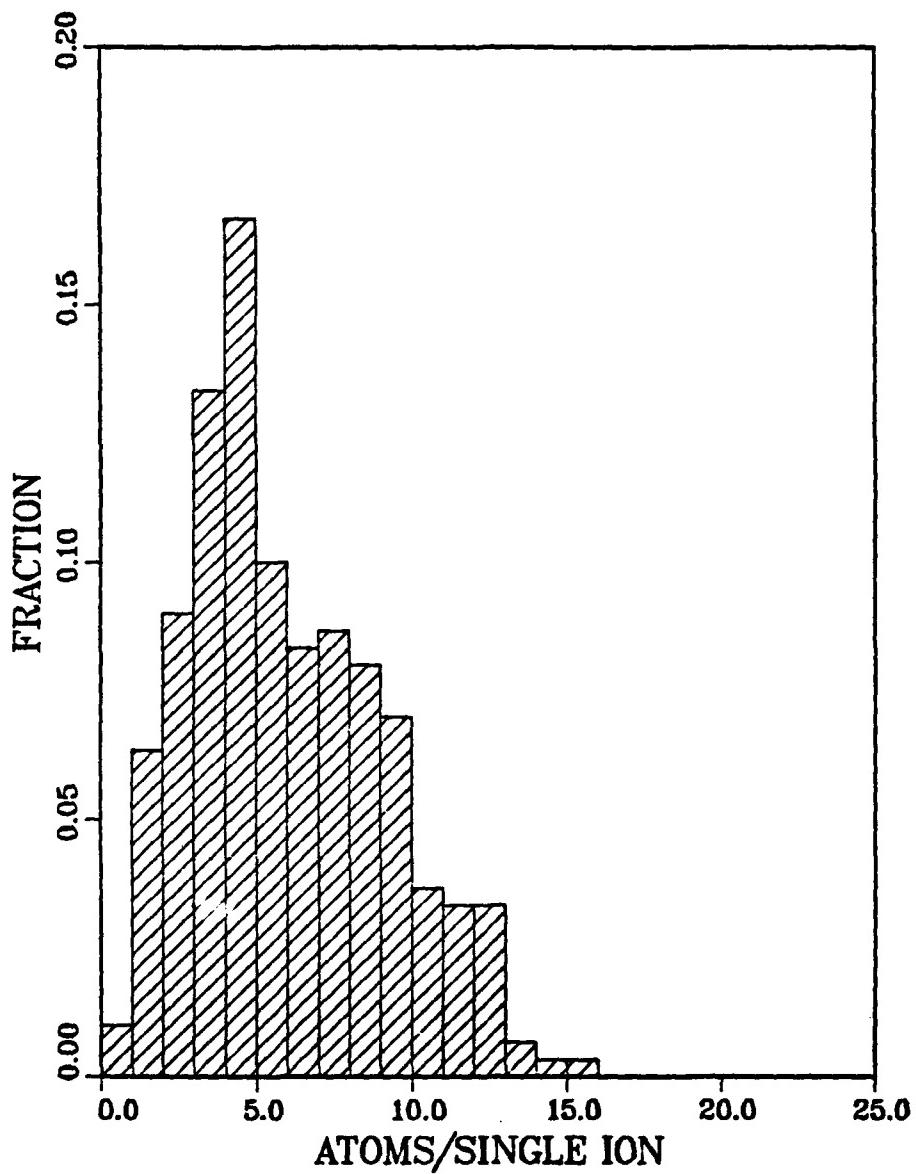


Figure 4 Atoms per Single Ion Distribution.

1.0 RH(111)/AR<111> WRMD DIFF TARG W/4 FROZEN
ATOMS/PER SINGLE ION

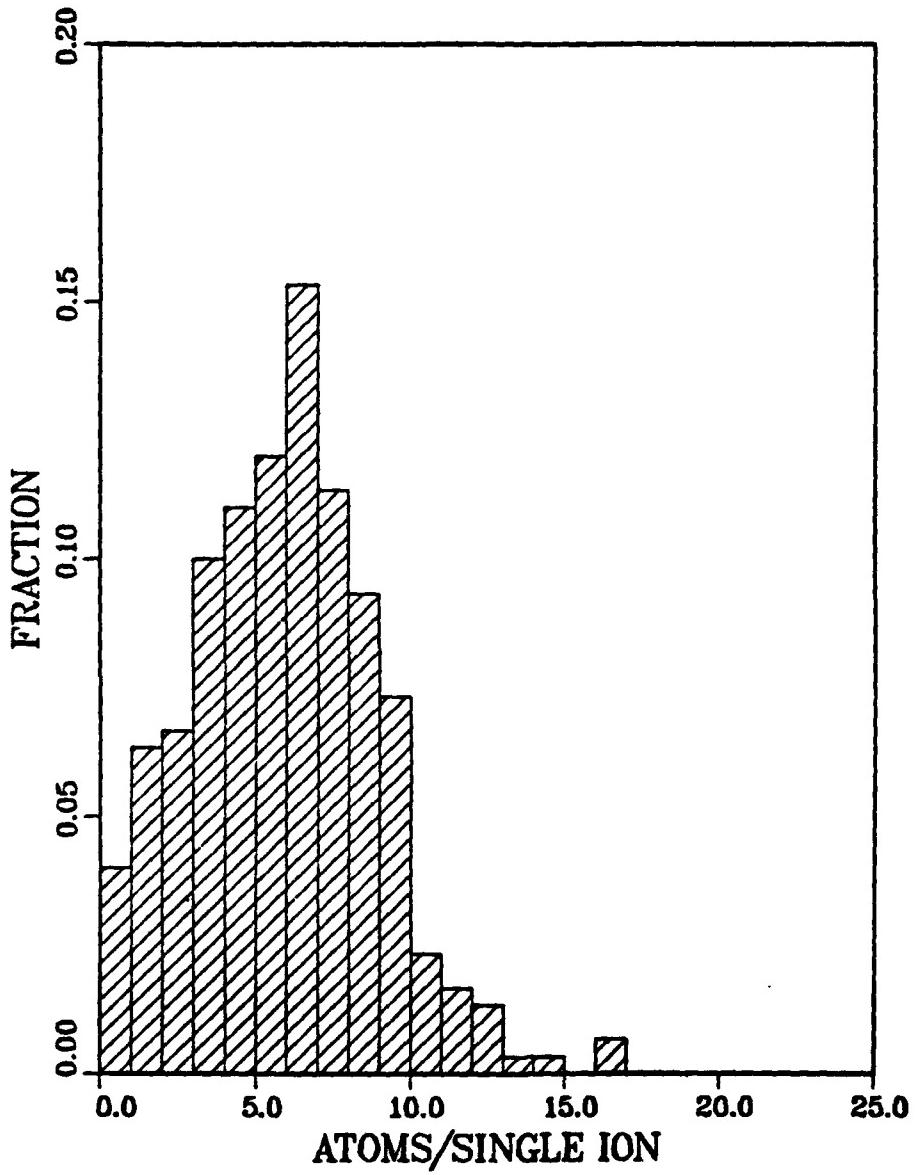


Figure 5 Atoms per Single Ion Distribution.

1.0 RH(111)/AR<111> SOLID TARGET
ATOM EJECTION TIME DISTRIBUTION

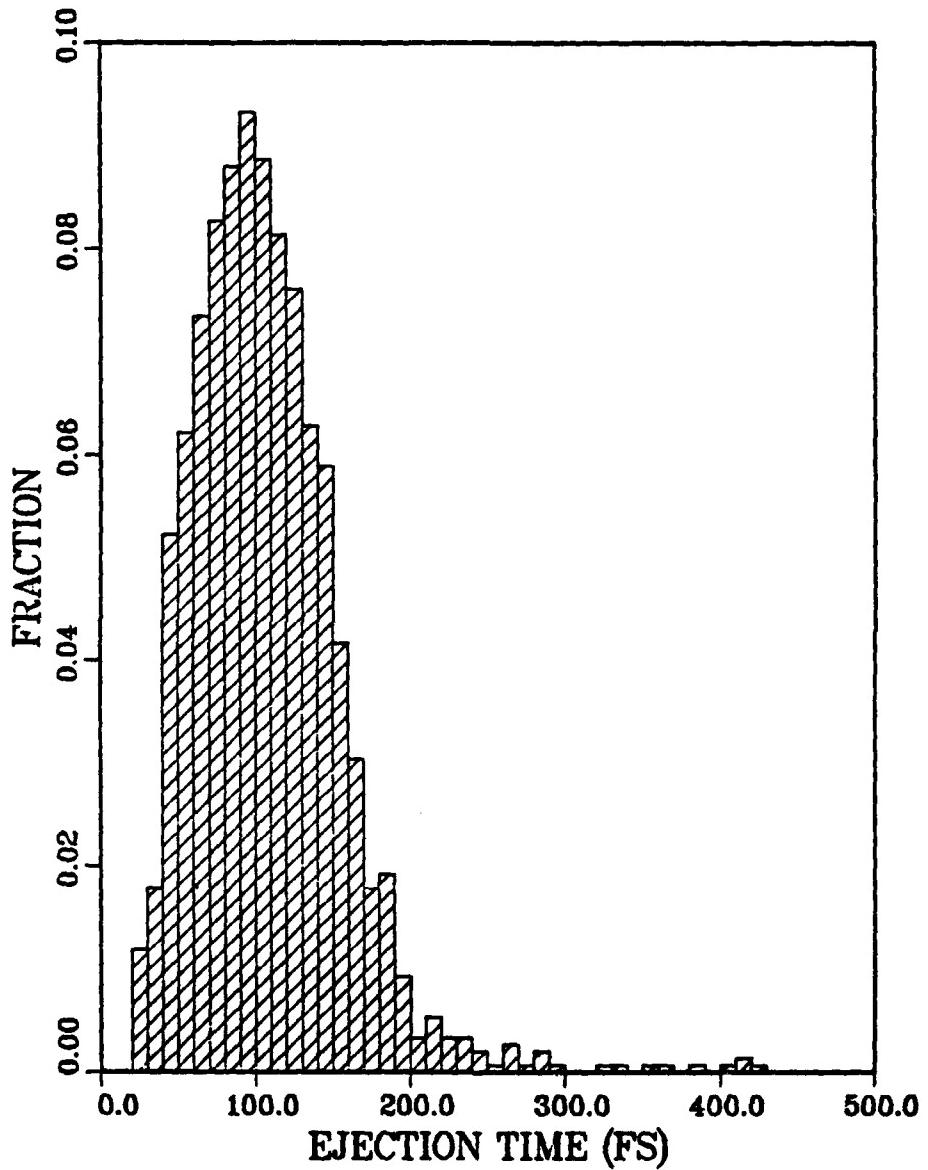


Figure 6 Atom Ejection Time Distribution.

1.0 RH(111)/AR<111> WARMED SINGLE TARGET
ATOM EJECTION TIME DISTRIBUTION

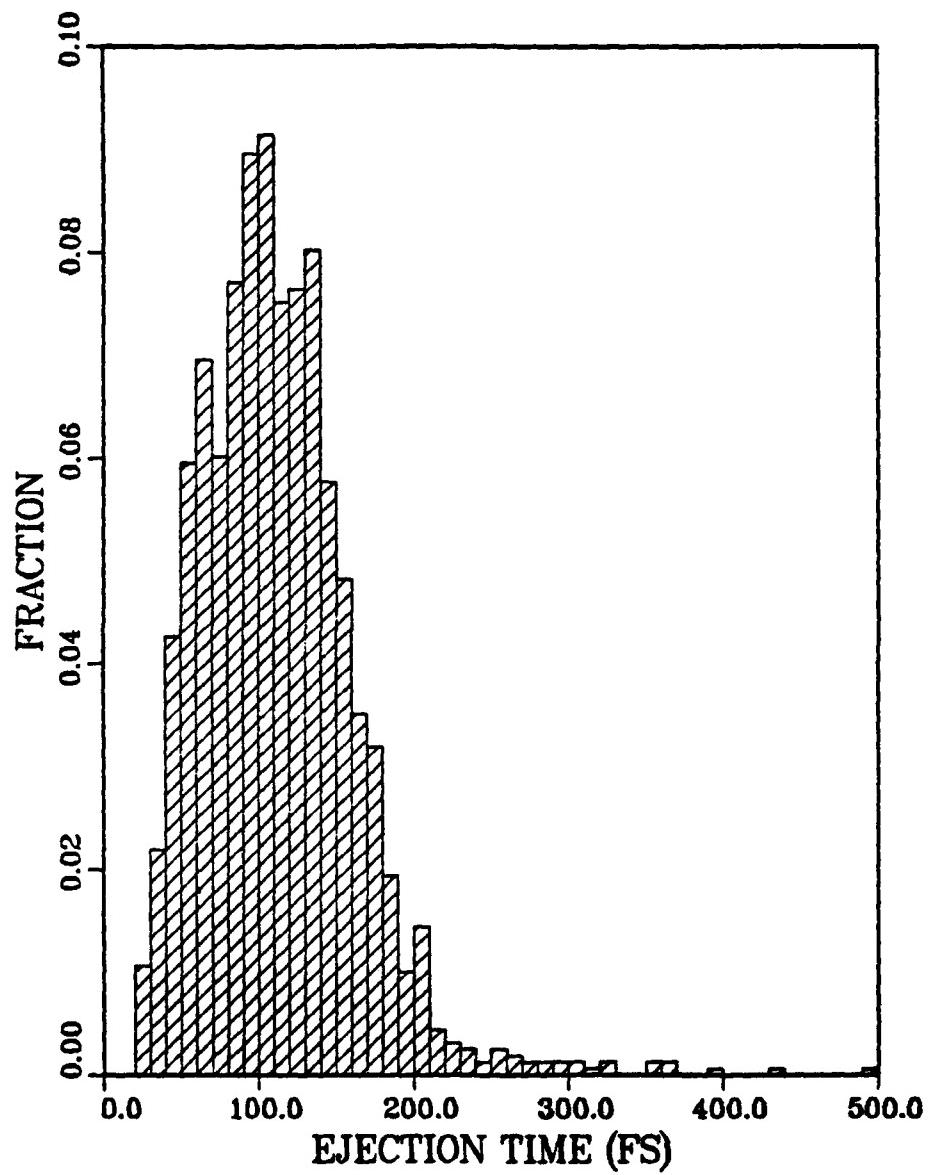


Figure 7 Atom Ejection Time Distribution.

1.0 RH(111)/AR<111> WRMD SING TARG W/4 FROZEN
ATOM EJECTION TIME DISTRIBUTION

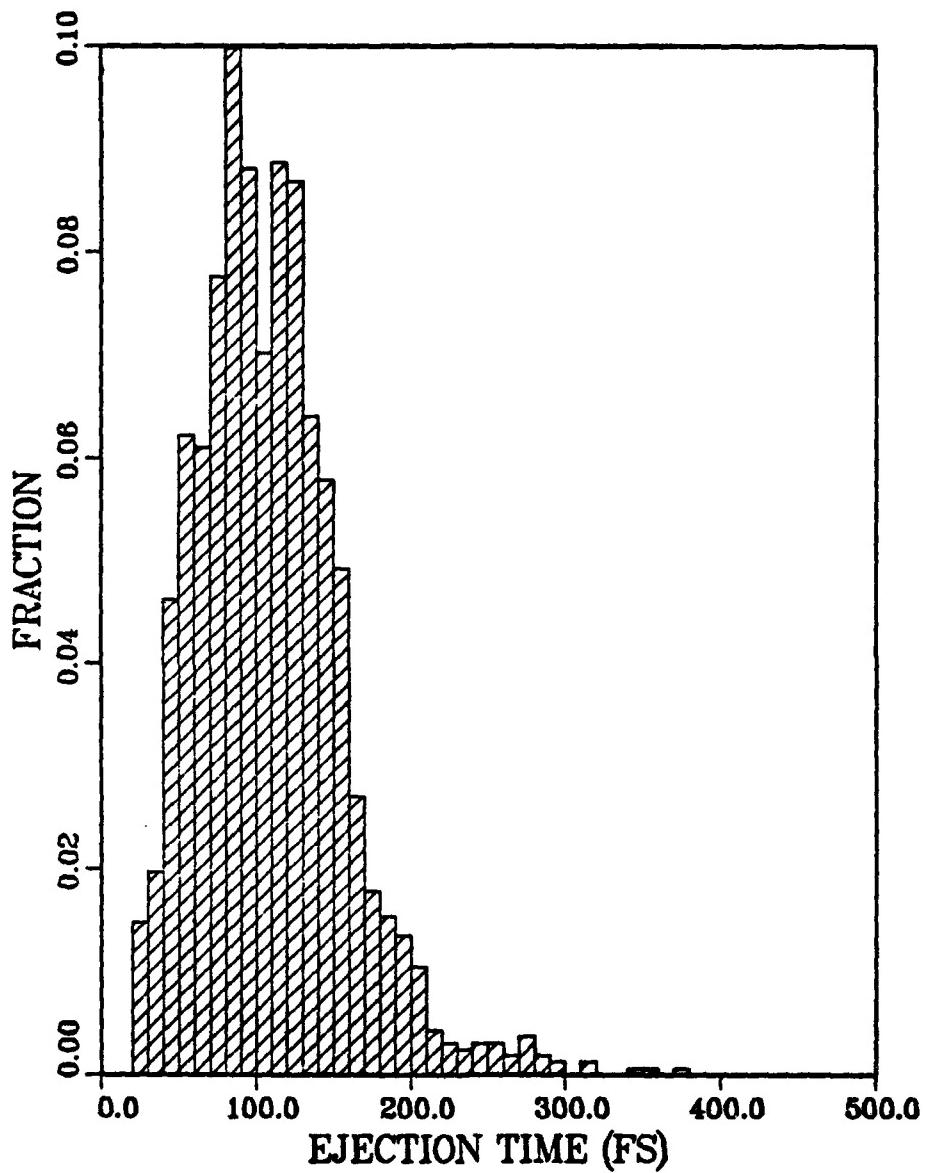


Figure 8 Atom Ejection Time Distribution.

1.0 RH(111)/AR<111> WARMED DIFFERENT TARGET
ATOM EJECTION TIME DISTRIBUTION

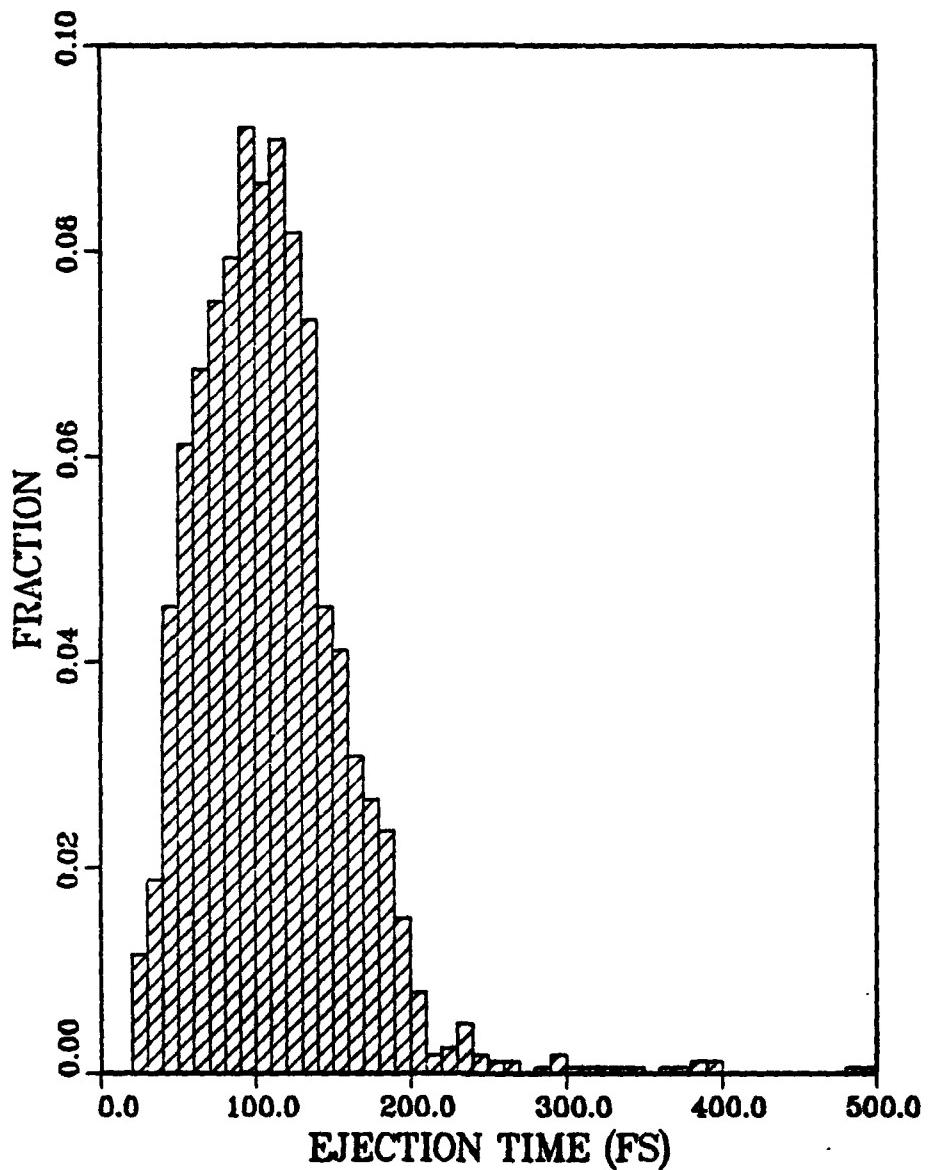


Figure 9 Atom Ejection Time Distribution.

1.0 RH(111)/AR<111> WRMD DIFF TARG W/4 FROZEN
ATOM EJECTION TIME DISTRIBUTION

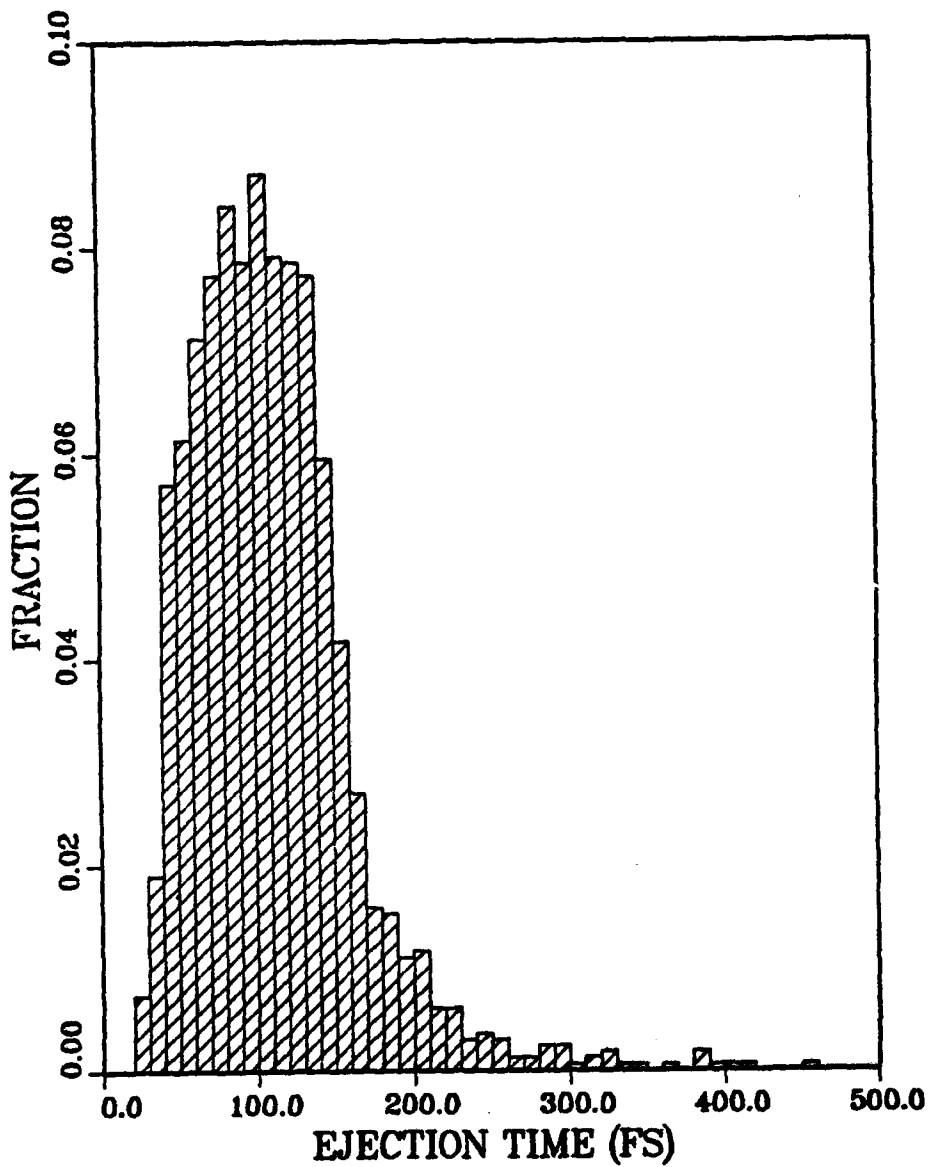


Figure 10 Atom Ejection Time Distribution.

1.0 RH(111)/AR<111> SOLID TARGET
EJECTION ANGLE DISTRIBUTION: ALL

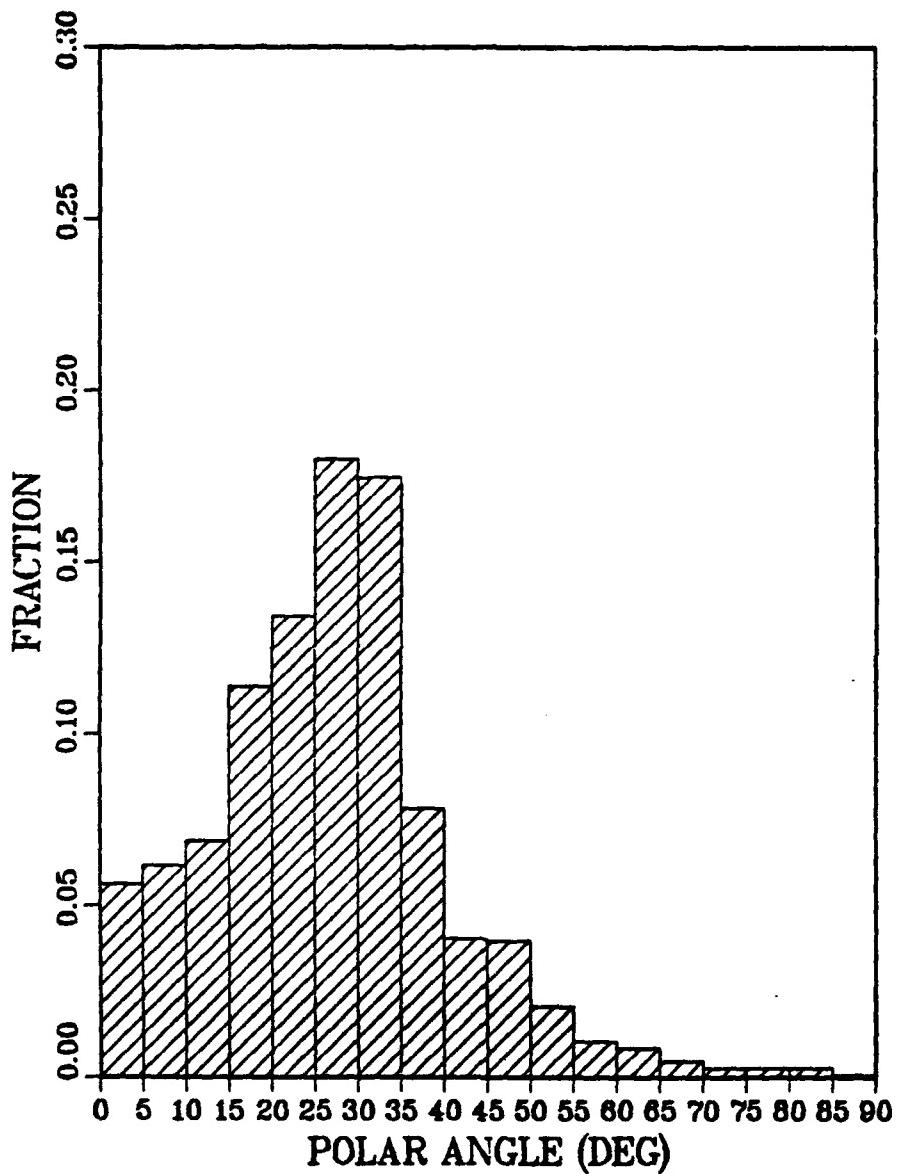


Figure 11 Ejected Atom Angular Distribution - All Atoms.

1.0 RH(111)/AR<111> WARMED SINGLE TARGET
EJECTION ANGLE DISTRIBUTION: ALL

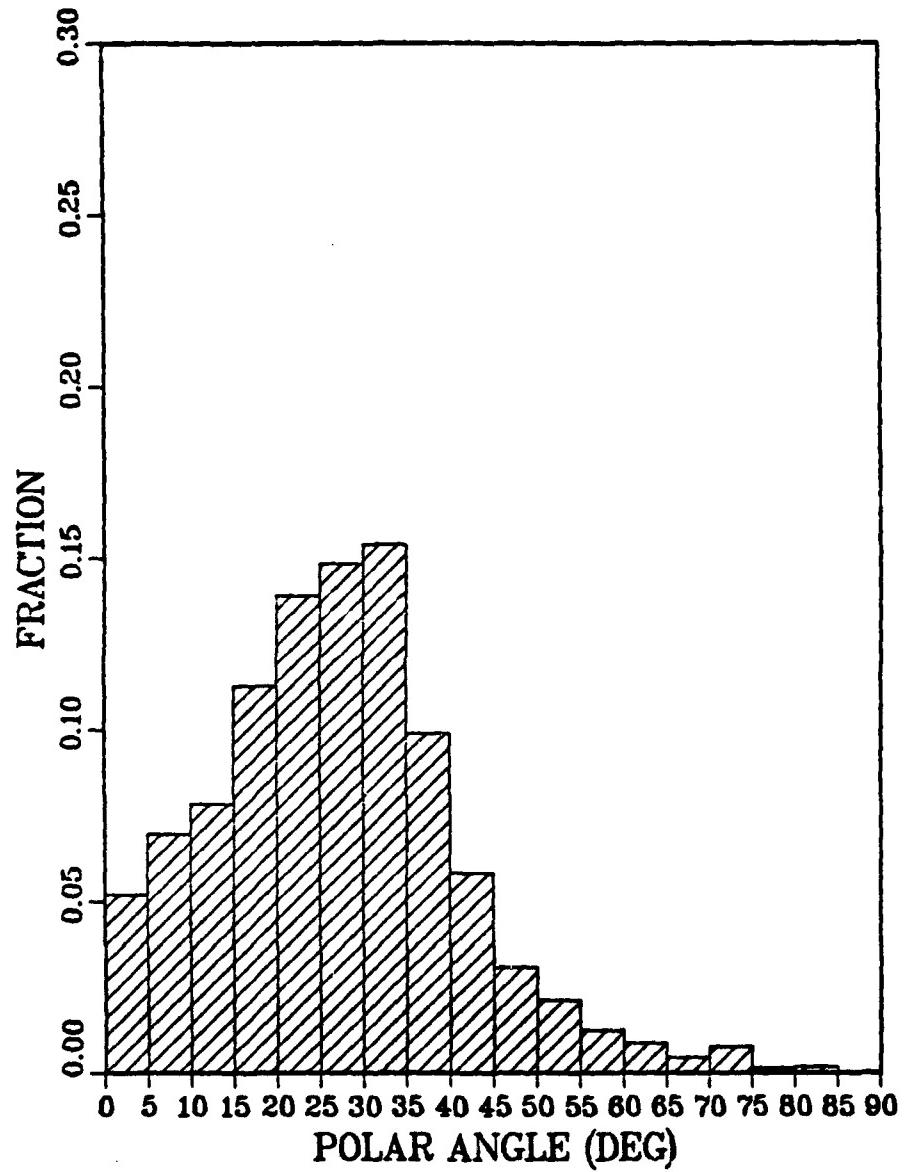


Figure 12 Ejected Atom Angular Distribution - All Atoms.

1.0 RH(111)/AR<111> WRMD SING TARG W/4 FROZEN
EJECTION ANGLE DISTRIBUTION: ALL

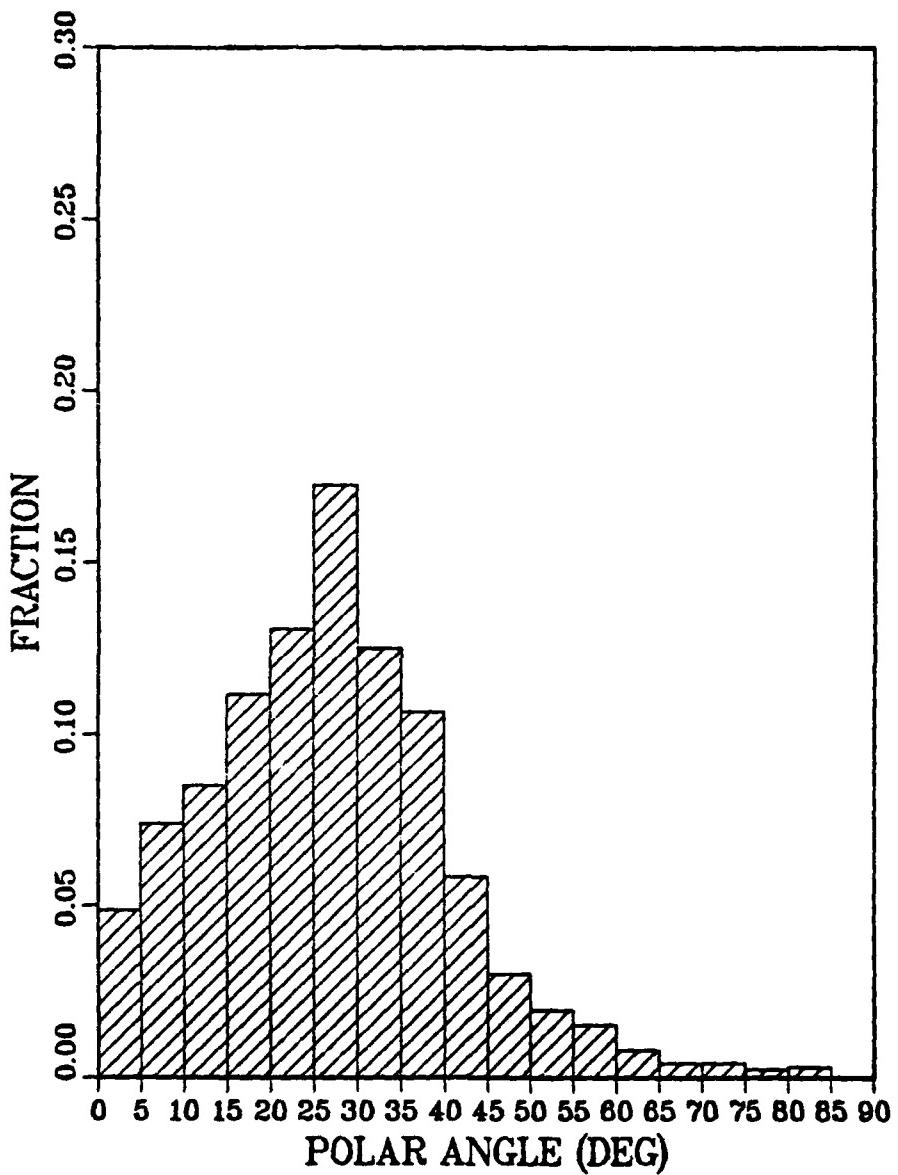


Figure 13 Ejected Atom Angular Distribution - All Atoms.

1.0 RH(111)/AR<111> WARMED DIFFERENT TARGET
EJECTION ANGLE DISTRIBUTION: ALL

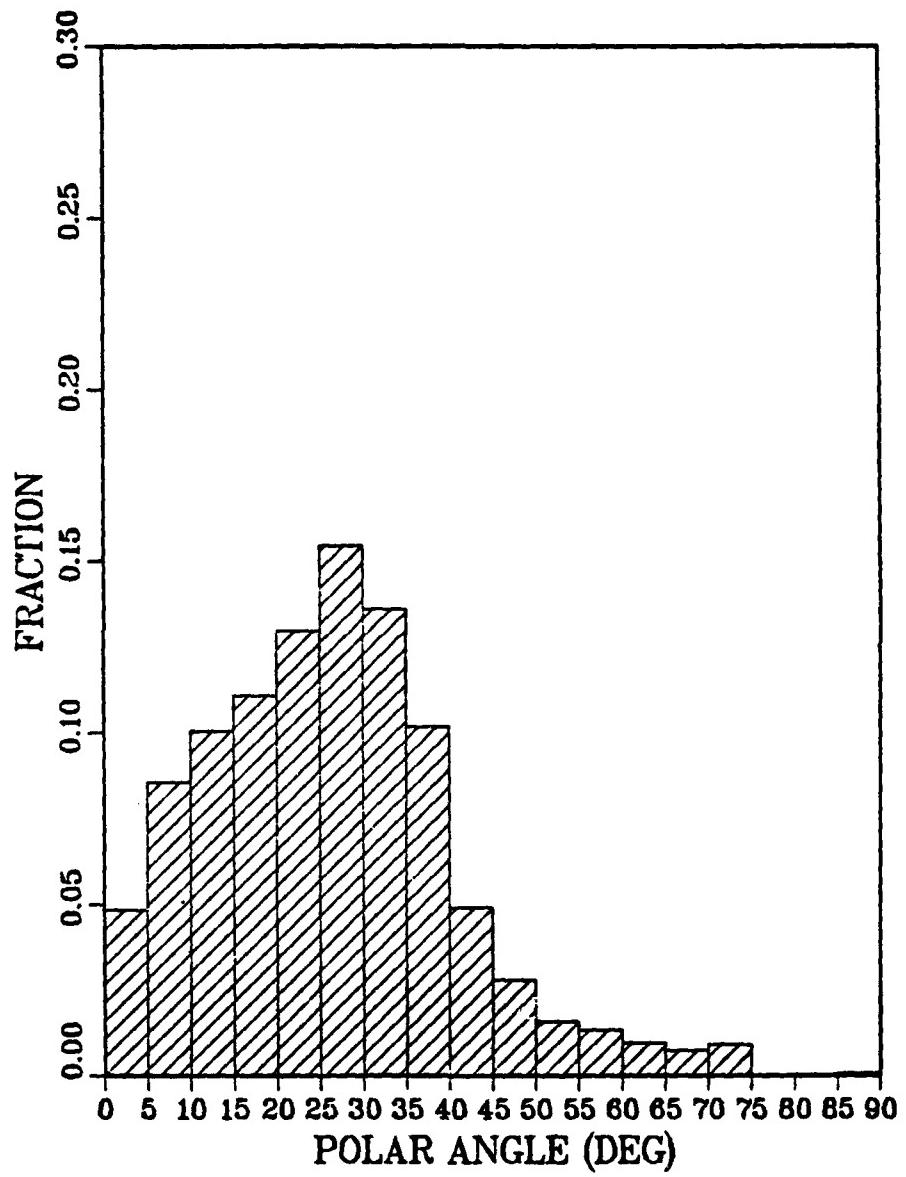


Figure 14 Ejected Atom Angular Distribution - All Atoms.

1.0 RH(111)/AR<111> WRMD DIFF TARG W/4 FROZEN
EJECTION ANGLE DISTRIBUTION: ALL

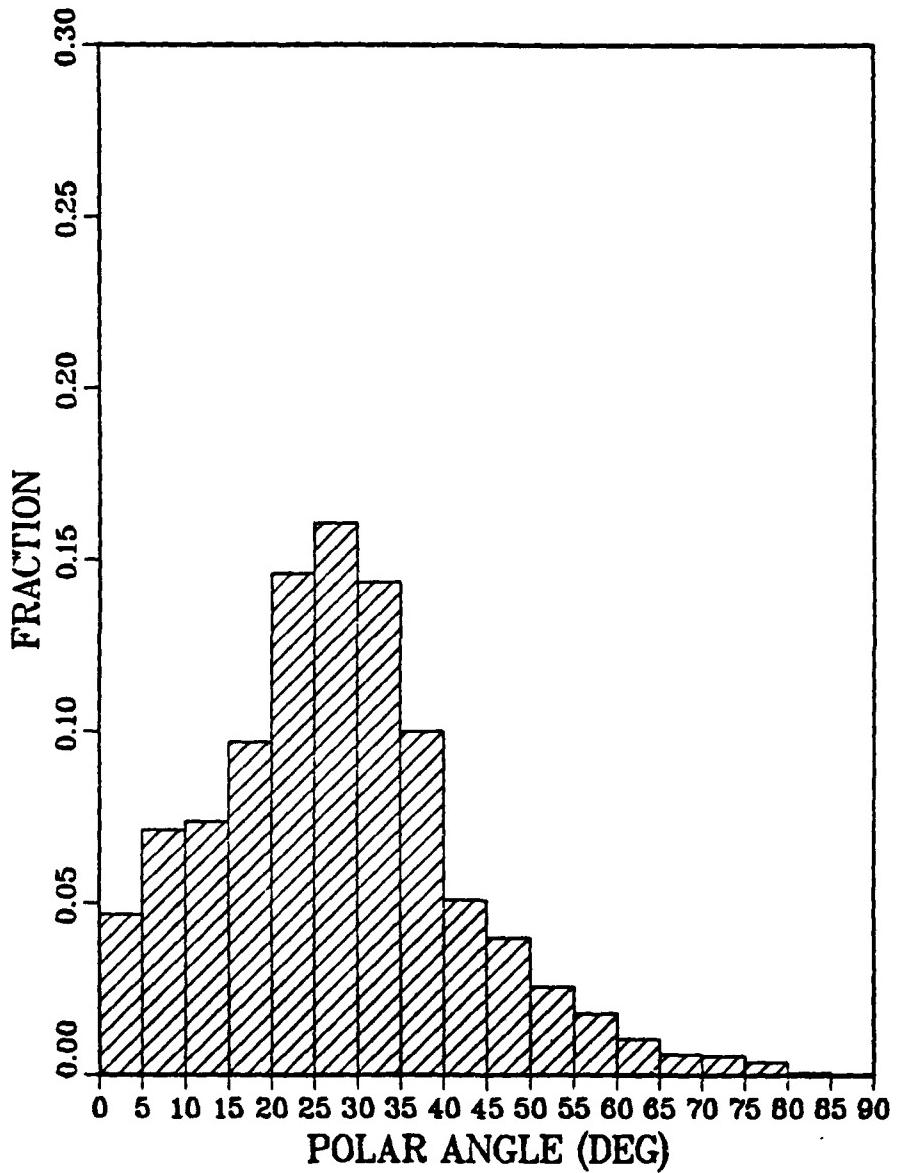


Figure 15 Ejected Atom Angular Distribution - All Atoms.

1.0 RH(111)/AR<111> SOLID TARGET
EJECTION ANGLE DISTRIBUTION > 20 eV

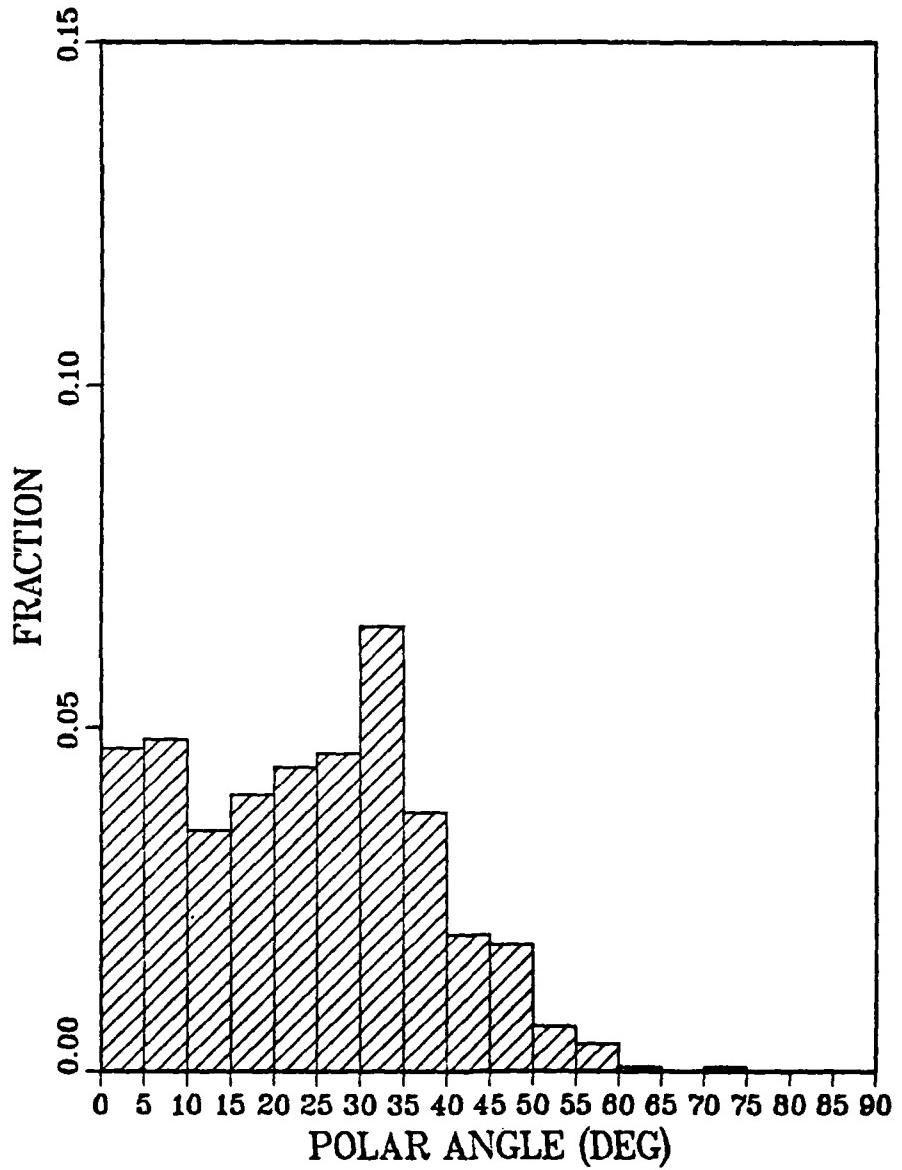
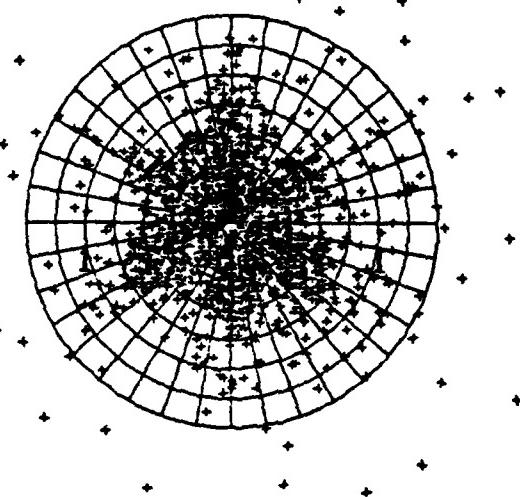


Figure 16 Ejected Atom Angular Distribution - Atoms > 20 eV.

1.0 RH(111)/AR<111> (19X6X16)

Solid Target



Warmed Single Target

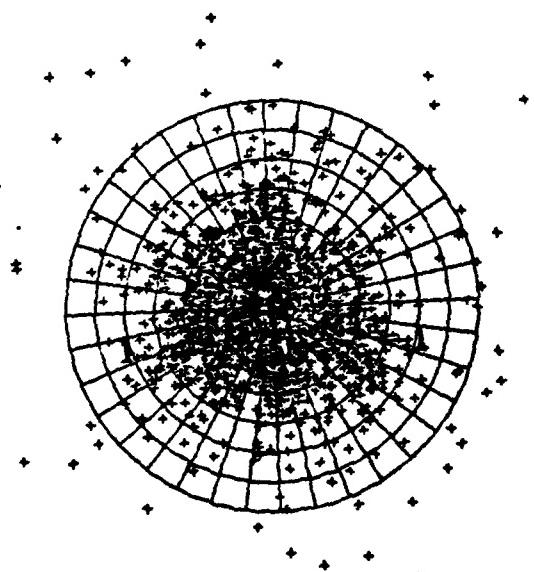
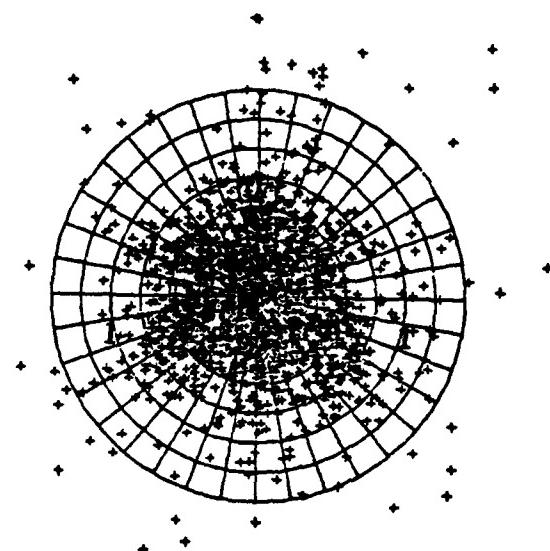


Figure 17 Spot Patterns.

1.0 RH(111)/AR<111> (19X6X16)

Warmed Single Target With Four Atoms Frozen



Warmed Different Target

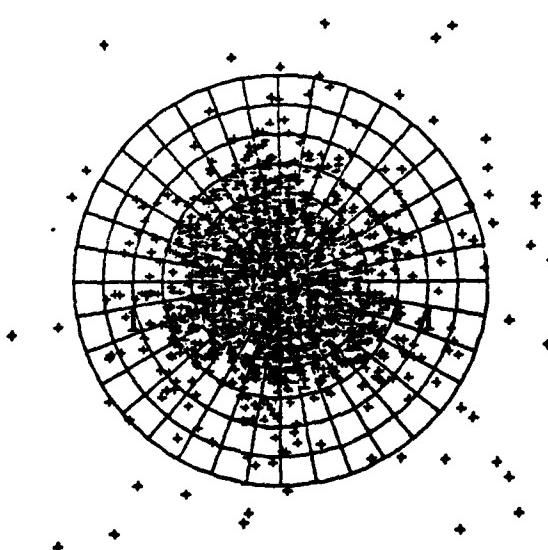


Figure 18 Spot Patterns.

1.0 RH(111)/AR<111> (19X6X16)

Warmed Different Target With Four Atoms Frozen

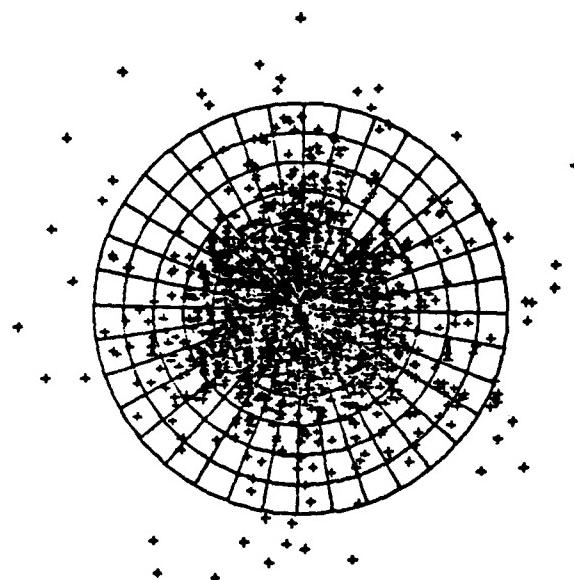


Figure 19 Spot Patterns.

1.0 RH(111)/AR<111> SOLID TARGET
ATOM YIELD PER IMPACT POINT

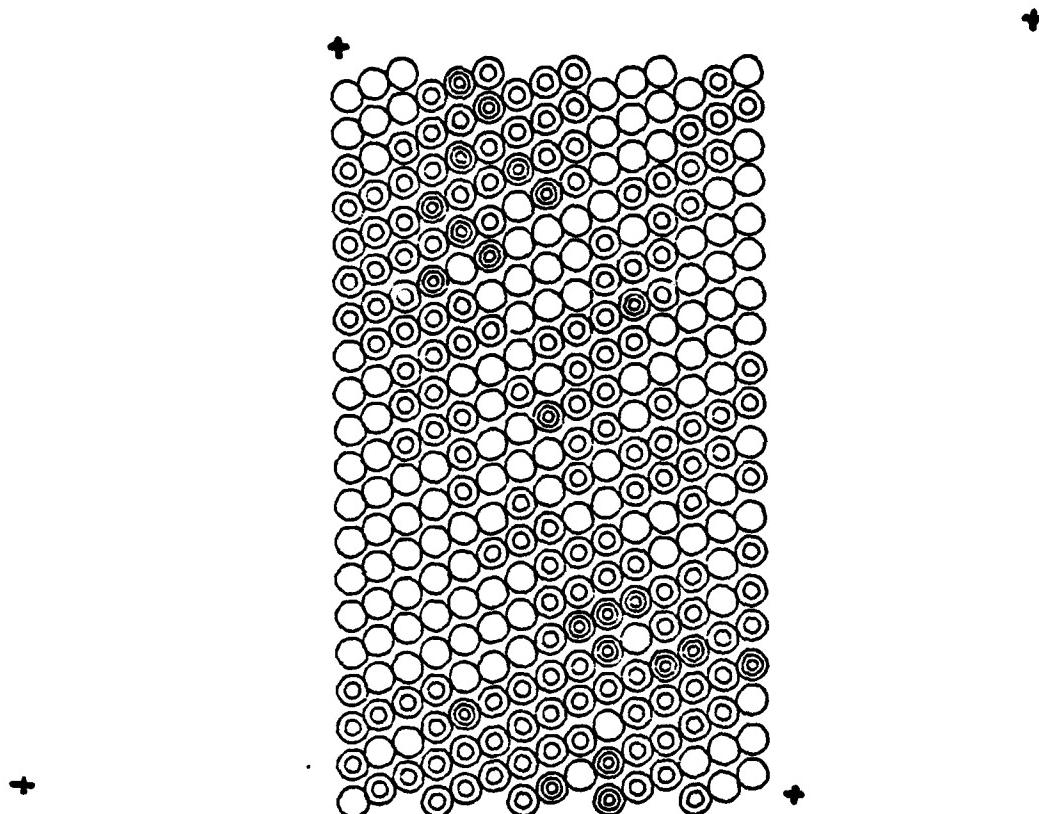


Figure 20 Atom Yield per Impact Point.

1.0 RH(111)/AR<111> WARMED SINGLE TARGET
ATOM YIELD PER IMPACT POINT

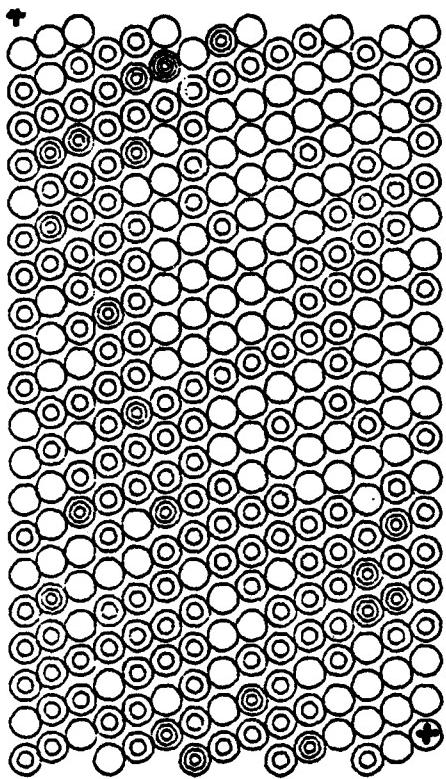


Figure 21 Atom Yield per Impact Point.

1.0 RH(111)/AR<111> WRMD SING TARG W/4 FROZEN
ATOM YIELD PER IMPACT POINT

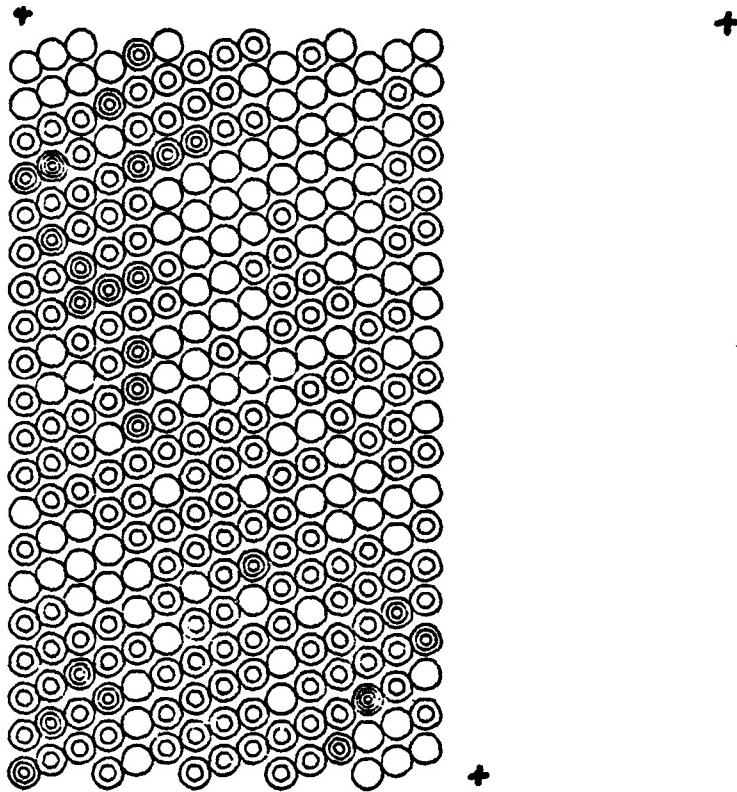


Figure 22 Atom Yield per Impact Point.

**1.0 RH(111)/AR<111> WARMED DIFFERENT TARGET
ATOM YIELD PER IMPACT POINT**

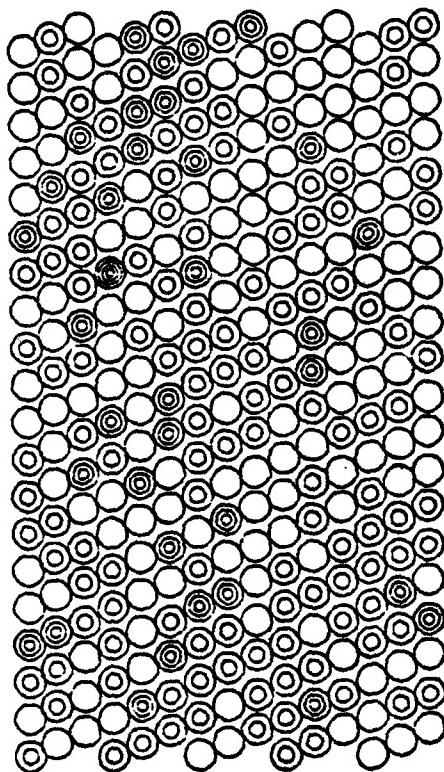


Figure 23 Atom Yield per Impact Point.

1.0 RH(111)/AR<111> WRMD DIFF TARG W/4 FROZEN
ATOM YIELD PER IMPACT POINT

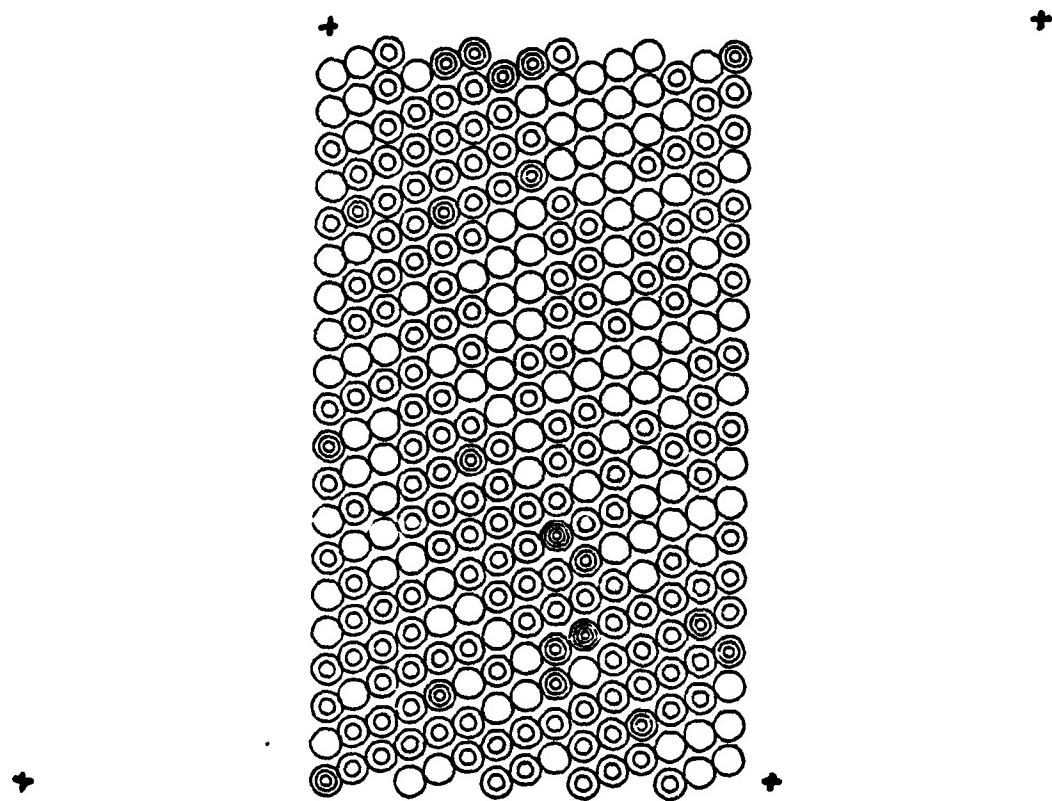


Figure 24 Atom Yield per Impact Point.

1.0 RH(111)/AR<111> SINGLE TRAJECTORY SET
ATOMS/PER SINGLE ION

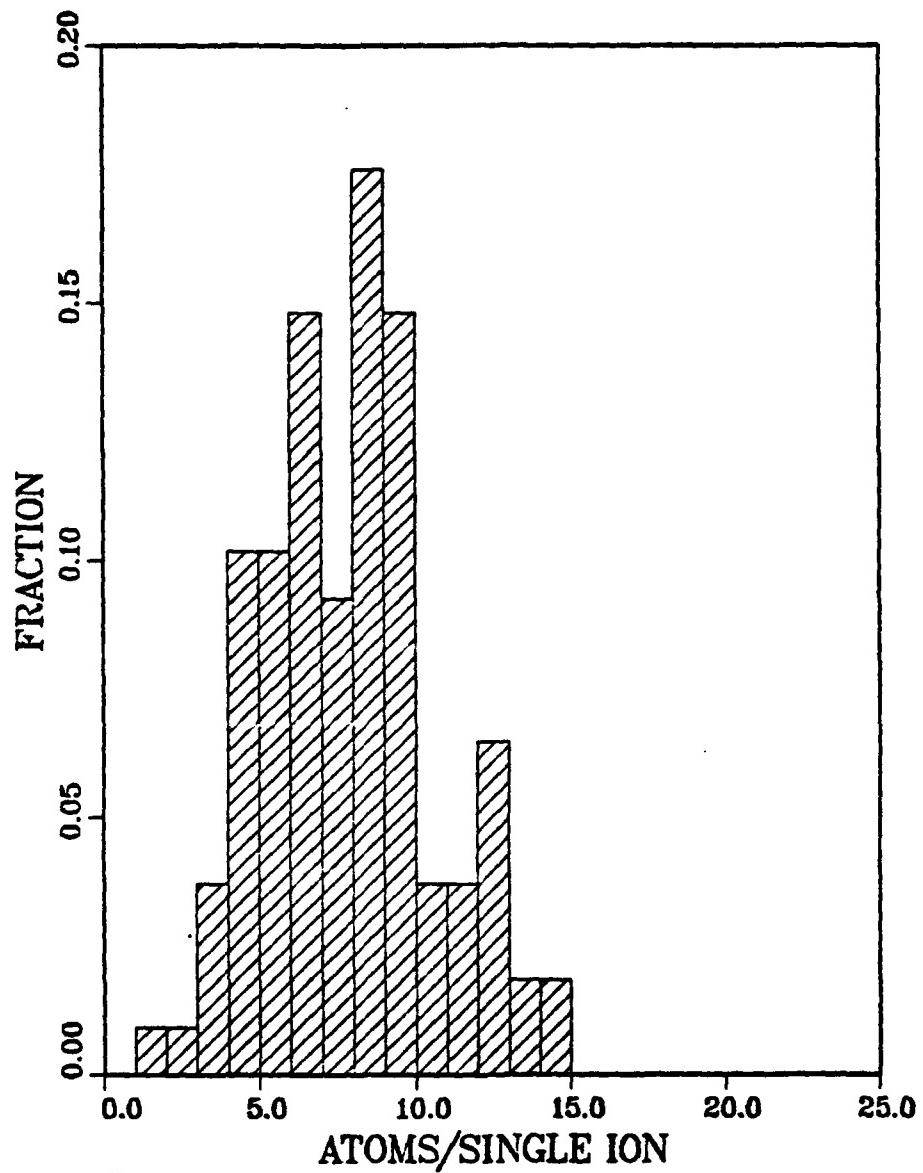


Figure 25 Atoms per Single Ion Distribution.

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